

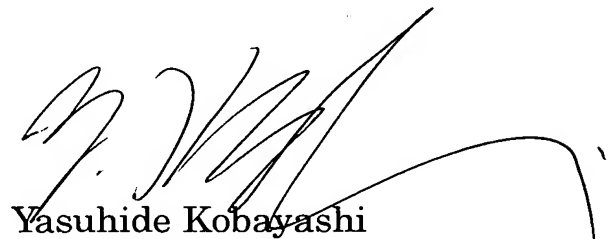


STATEMENT

I, Yasuhide KOBAYASHI—of ARK Mori Building, 13F, 12-32, Akasaka 1-chome, Minato-ku, Tokyo 107-6013 Japan—hereby state that I have a thorough knowledge of the English and Japanese languages and that the attached document is an accurate English translation of the Priority document of Japanese Patent Application No. Hei. 11-304206 filed October 26, 1999, upon which the present application claims a priority.

Declared at Tokyo, Japan

This 21th day of July, 2005



Yasuhide Kobayashi



PATENT OFFICE
JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy of the following application as filed with this Office.

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Application Number: Japanese Patent Application
No. Hei. 11-304206

Applicant: FUJI PHOTO FILM CO., LTD.

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Commissioner, Patent Office Kouzo Oikawa (sealed)

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(Inventor)

 (Address or c/o Fuji Photo Film Co., Ltd., 210,
 Residence) Nakanuma, Minami Ashigara-shi,
 Kanagawa, Japan

 (Name) Tatsuya Igarashi

(Applicant for Patent)

 (Identification No.) 000005201

 (Appellation) FUJI PHOTO FILM CO., LTD.

(AGENT)

 (Identification No.) 100073874

 (Attorney)

 (Name) Taira Hagino

 (Phone Number) 03-5561-3990

(Selected AGENT)

 (Identification No.) 100066429

 (Attorney)

 (Name) Toshio Fukazawa

 (Phone Number) 03-5561-3990

(Selected AGENT)

 (Identification No.) 100093573

 (Attorney)

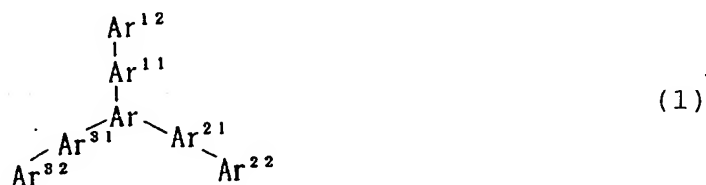
(Name) Zenichi Soeda
(Phone Number) 03-5561-3990
(Selected AGENT)
(Identification No.) 100105474
(Attorney)
(Name) Hironori Honda
(Phone Number) 03-5561-3990
(Selected AGENT)
(Identification No.) 100090343
(Attorney)
(Name) Yuriko Kuriu
(Phone Number) 03-5561-3990
(Indication of Fee)
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[Designation of Document] Specification

[Title of the Invention] AROMATIC CONDENSED-RING COMPOUND,
LIGHT EMITTING DEVICE MATERIAL AND LIGHT EMITTING DEVICE USING
THE SAME

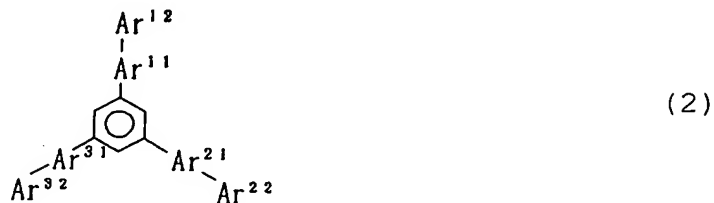
[Claims]

1. A light emitting device material comprising a compound represented by the following formula (1):



wherein Ar¹¹, Ar²¹ and Ar³¹ represent an arylene group, Ar¹², Ar²² and Ar³² represent a substituent group or a hydrogen atom, provided that at least one of Ar¹¹, Ar²¹, Ar³¹, Ar¹², Ar²² and Ar³² has a condensed-ring aryl structure or a condensed-ring heteroaryl structure, and Ar represents an arylene group or a heteroarylene group.

2. A light emitting device material comprising a compound represented by the following formula (2):



wherein Ar¹¹, Ar²¹ and Ar³¹ represent an arylene group, and Ar¹², Ar²² and Ar³² represent a substituent group or a hydrogen atom, provided that at least one of Ar¹¹, Ar²¹, Ar³¹, Ar¹², Ar²² and Ar³² has a condensed-ring aryl structure or a condensed-ring heteroaryl structure.

3. A light emitting device material as described in claim 1 or 2, wherein Ar¹², Ar²² and Ar³² are an aryl group.

4. A light emitting device material as described in claim 1 or 2, wherein Ar¹², Ar²² and Ar³² are a hydrogen atom.

5. A light emitting device material as described in claim 1, 2, 3 or 4, wherein at least one of Ar¹¹, Ar²¹, Ar³¹, Ar¹², Ar²² and Ar³² has a condensed-ring aryl structure in which at least 3 rings are condensed, or a condensed-ring heteroaryl structure in which at least 3 rings are condensed.

6. A light emitting device material as described in claim 1, 2, 3, 4 or 5, wherein Ar¹¹, Ar²¹ and Ar³¹ are a condensed-ring arylene group.

7. A light emitting device material as described in claim 1, 2, 3, 4 or 5, wherein Ar¹¹, Ar²¹ and Ar³¹ are an condensed-ring arylene group in which at least 3 rings are condensed.

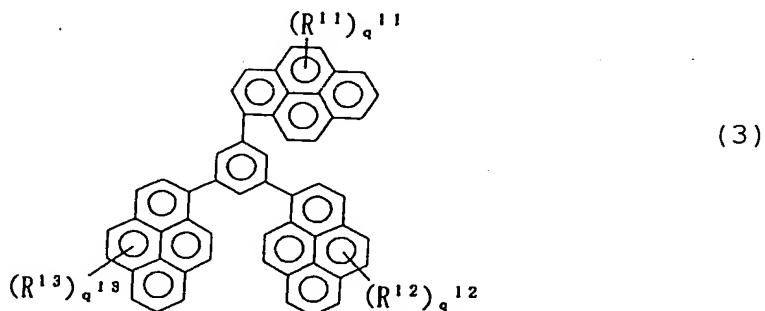
8. A light emitting device material as described in claim 1, 2, 3, 5, 6 or 7, wherein Ar¹², Ar²² and Ar³² are a condensed-ring aryl group.

9. A light emitting device material as described in

claim 1, 2, 3, 5, 6 or 7, wherein Ar^{12} , Ar^{22} and Ar^{32} are a condensed-ring aryl group in which at least 3 rings are condensed.

10. A light emitting device material as described in claim 1, 2, 3, 4, 5, 6, 7, 8 or 9, wherein said compound is constituted of carbon and hydrogen atoms alone.

11. A compound represented by the following formula (3):



wherein R^{11} , R^{12} and R^{13} represent a substituent group, and q^{11} , q^{12} and q^{13} represent an integer of 0 to 9.

12. A light emitting device material comprising the compound of formula (3) defined in claim 11.

13. A light emitting device having between a pair of electrodes a luminescent layer or a plurality of thin organic compound layers including a luminescent layer, wherein at least one layer is a layer comprising at least one light emitting device material as described in claim 1, 2, 3, 4, 5, 6, 7, 8, 9, 10 or 12.

14. An organic light emitting device as described in

claim 13, wherein the layer comprising the light emitting device material as described in claim 1, 2, 3, 4, 5, 6, 7, 8, 9, 10 or 12 is a layer formed in a coating process.

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Belongs]

The present invention relates to an aromatic condensed-ring compound, a material used for light emitting devices capable of luminescing through conversion of electric energy into light, and a light emitting device using such a material. In particular, the invention is concerned with a light emitting device suitably used in the areas of indicators, displays, backlight, electrophotography, illumination sources, recording light sources, exposure illuminants, reading light sources, beacons, signboards and interiors.

[0002]

[Prior Art]

Nowadays, lively researches and developments in various display devices are being made. Of such devices, the organic electric-field luminescent (EL) devices are receiving particular attention as promising display devices because they can emit light of high intensity under low voltage. For instance, the EL devices having organic thin films formed by vapor deposition of organic compounds are known (Applied Physics Letters, 51, p.913 (1987)). The organic EL devices described

in the literature cited above employ tris(8-hydroxyquinolinato)aluminum complex (Alq) as an electron transporting material and is structured to superimpose a layer of positive hole transporting material (an amine compound) on the layer of electron transporting material, and thereby show substantial improvements in their luminescent characteristics over conventional single-layer devices.

In recent years, the application of organic EL devices to full-color display has been examined actively. In order to develop high-performance full-color display, it is necessary to improve on the characteristics of conventional blue, green and red light emitting devices respectively. As to the blue light emitting devices, for instance, the distyrylarylene compounds (DPVBi) described in a book entitled "Organic EL devices and Forefront of Industrialization thereof", at page 38 (published by NTS Co.) have been studied extensively, but they have problems with color purity, durability, luminous intensity and efficiency, and so further improvements in these characteristics have been expected.

[0003]

Although the organic light emitting devices comprising organic materials laminated by vacuum evaporation have really succeeded in luminescence of high luminance, the use of coating methods for manufacturing devices have advantages over that of vacuum evaporation methods from the viewpoints of simplicity

of the manufacturing process, workability and ease in achievement of large-area luminescence. However, the devices, especially blue light emitting devices, manufactured according to conventional coating methods are inferior to those manufactured using vapor deposition methods in luminous intensity and luminous efficiency. Therefore, the development of novel blue luminescent materials has been awaited.

[0004]

[Problems that the Invention is to Solve]

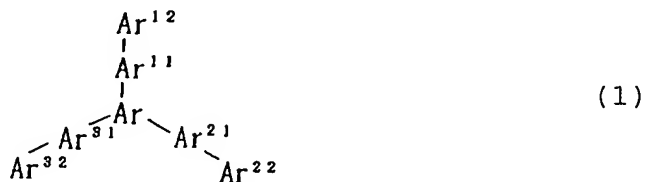
An object of the invention is to provide a light emitting device having satisfactory luminescence characteristics and a material enabling a light emitting device to have such characteristics.

[0005]

The object was attained with the following means.

(1) A light emitting device material comprising a compound represented by the following formula (1):

[0006]



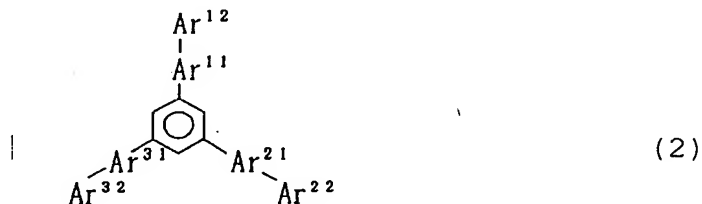
[0007]

wherein Ar^{11} , Ar^{21} and Ar^{31} represent an arylene group, Ar^{12} , Ar^{22}

and Ar³² represent a substituent group or a hydrogen atom, provided that at least one of Ar¹¹, Ar²¹, Ar³¹, Ar¹², Ar²² and Ar³² has a condensed-ring aryl structure or a condensed-ring heteroaryl structure, and Ar represents an arylene group or a heteroarylene group.

(2) A light emitting device material comprising a compound represented by the following formula (2):

[0008]



[0009]

wherein Ar¹¹, Ar²¹ and Ar³¹ represent an arylene group, and Ar¹², Ar²² and Ar³² represent a substituent group or a hydrogen atom, provided that at least one of Ar¹¹, Ar²¹, Ar³¹, Ar¹², Ar²² and Ar³² has a condensed-ring aryl structure or a condensed-ring heteroaryl structure.

(3) A light emitting device material as described in (1) or (2), wherein Ar¹², Ar²² and Ar³² are an aryl group.

(4) A light emitting device material as described in (1) or (2), wherein Ar¹², Ar²² and Ar³² are a hydrogen atom.

(5) A light emitting device material as described in (1), (2), (3) or (4), wherein at least one of Ar¹¹, Ar²¹, Ar³¹, Ar¹², Ar²² and Ar³² has a condensed-ring aryl structure in which

at least 3 rings are condensed, or a condensed-ring heteroaryl structure in which at least 3 rings are condensed.

(6) A light emitting device material as described in (1), (2), (3), (4) or (5), wherein Ar^{11} , Ar^{21} and Ar^{31} are a condensed-ring arylene group.

(7) A light emitting device material as described in (1), (2), (3), (4) or (5), wherein Ar^{11} , Ar^{21} and Ar^{31} are an condensed-ring arylene group in which at least 3 rings are condensed.

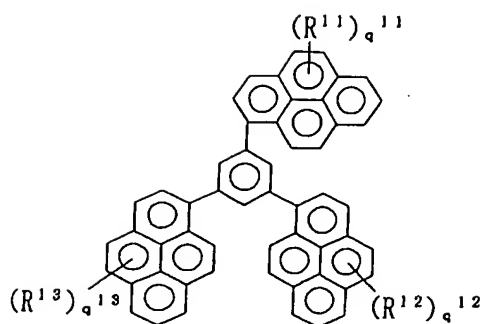
(8) A light emitting device material as described in (1), (2), (3) or (5), wherein Ar^{12} , Ar^{22} and Ar^{32} are a condensed-ring aryl group.

(9) A light emitting device material as described in (1), (2), (3) or (5), wherein Ar^{12} , Ar^{22} and Ar^{32} are a condensed-ring aryl group in which at least 3 rings are condensed.

(10) A light emitting device material as described in (1), (2), (3), (4), (5), (6), (7), (8) or (9), wherein the compound is constituted of carbon and hydrogen atoms alone.

(11) A compound represented by the following formula (3):

[0010]



[0011]

wherein R^{11} , R^{12} and R^{13} represent a substituent group, and q^{11} , q^{12} and q^{13} represent an integer of 0 to 9.

(12) A light emitting device material comprising a compound of formula (3) defined in (11).

(13) A light emitting device having between a pair of electrodes a luminescent layer or a plurality of thin organic compound layers including a luminescent layer, wherein at least one layer is a layer comprising at least one light emitting device material as described in (1), (2), (3), (4), (5), (6), (7), (8), (9), (10) or (12).

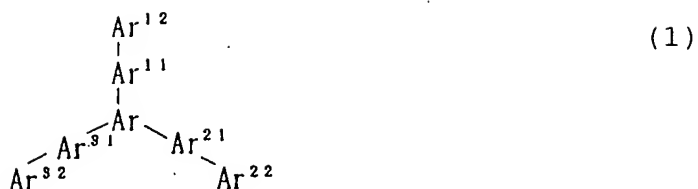
(14) An organic light emitting device as described in (13), wherein the layer comprising a light emitting device material as described in (1), (2), (3), (4), (5), (6), (7), (8), (9), (10) or (12) is a layer formed in a coating process.

[0012]

[Embodiment of Carrying Out the Invention]

The invention is described below in detail.

[0013]



[0014]

The formula (1) representing compounds according to the invention are illustrated first.

Ar^{11} , Ar^{21} and Ar^{31} represent an arylene group. The suitable number of carbon atoms contained in the arylene group is from 6 to 30, preferably from 6 to 20, and particularly preferably from 6 to 16. Examples of such an arylene group include a phenylene group, a naphthylene group, an anthracenylene group, a phenanthrenylene group, a pyrenylene group, a perylenylene group, a fluorenylene group, a biphenylene group, a terphenylene group, a rubrenylene group, a chrysenylene group, a triphenylene group, a benzanthracenylene group, a benzophenanthrenylene group and a diphenylanthracenylene group. These arylene groups each may have a substituent group.

[0015]

Examples of a substituent group on an arylene group include alkyl groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 10 carbon atoms, such as methyl, ethyl, iso-propyl, tert-butyl, n-octyl, n-decyl, n-hexadecyl, cyclopropyl, cyclopentyl and cyclohexyl groups), alkenyl groups (containing preferably 2 to 30 carbon atoms, more preferably 2 to 20 carbon

atoms, particularly preferably 2 to 10 carbon atoms, such as vinyl, allyl, 2-butenyl and 3-pentenyl groups), alkynyl groups (containing preferably 2 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, particularly preferably 2 to 10 carbon atoms, such as propargyl and 3-pentynyl groups), aryl groups (containing preferably 6 to 30 carbon atoms, more preferably 6 to 20 carbon atoms, particularly preferably 6 to 12 carbon atoms, such as phenyl, p-methylphenyl, naphthyl and anthranyl groups), amino groups (containing preferably 0 to 30 carbon atoms, more preferably 0 to 20 carbon atoms, particularly preferably 0 to 10 carbon atoms, such as amino, methylamino, dimethylamino, diethylamino, dibenzylamino, diphenylamino and ditolylamino groups), alkoxy groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 10 carbon atoms, such as methoxy, ethoxy, butoxy and 2-ethylhexyloxy groups), aryloxy groups (containing preferably 6 to 30 carbon atoms, more preferably 6 to 20 carbon atoms, particularly preferably 6 to 12 carbon atoms, such as phenyloxy, 1-naphthyloxy and 2-naphthyloxy groups), heteroaryloxy groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon atoms, such as pyridyloxy, pyrazyloxy, pyrimidyloxy and quinolyloxy groups), acyl groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon atoms, such as

acetyl, benzoyl, formyl and pivaloyl groups), alkoxycarbonyl groups (containing preferably 2 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, particularly preferably 2 to 12 carbon atoms, such as methoxycarbonyl and ethoxycarbonyl groups), aryloxy carbonyl groups (containing preferably 7 to 30 carbon atoms, more preferably 7 to 20 carbon atoms, particularly preferably 7 to 12 carbon atoms, such as phenyloxycarbonyl group), acyloxy groups (containing preferably 2 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, particularly preferably 2 to 10 carbon atoms, such as acetoxy and benzoyloxy groups), acylamino groups (containing preferably 2 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, particularly preferably 2 to 10 carbon atoms, such as acetyl amino and benzoylamino groups), alkoxycarbonylamino groups (containing preferably 2 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, particularly preferably 2 to 12 carbon atoms, such as methoxycarbonylamino group), aryloxy carbonylamino groups (containing preferably 7 to 30 carbon atoms, more preferably 7 to 20 carbon atoms, particularly preferably 7 to 12 carbon atoms, such as phenyloxycarbonylamino group), sulfonylamino groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon atoms, such as methanesulfonylamino and benzenesulfonylamino groups), sulfamoyl groups (containing preferably 0 to 30 carbon atoms, more preferably 0 to 20 carbon

atoms, particularly preferably 0 to 12 carbon atoms, such as sulfamoyl, methylsulfamoyl, dimethylsulfamoyl and phenylsulfamoyl groups), carbamoyl groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon atoms, such as carbamoyl, methylcarbamoyl, diethylcarbamoyl and phenylcarbamoyl groups), alkylthio groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon atoms, such as methylthio and ethylthio groups), arylthio groups (containing preferably 6 to 30 carbon atoms, more preferably 6 to 20 carbon atoms, particularly preferably 6 to 12 carbon atoms, such as phenylthio group), heteroarylthio groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon atoms, such as pyridylthio, 2-benzimidazolylthio, 2-benzoxazolylthio and 2-benzothiazolylthio groups), sulfonyl groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon atoms, such as mesyl and tosyl groups), sulfinyl groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon atoms, such as methanesulfinyl and benzenesulfinyl groups), ureido groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon

atoms, such as ureido, methylureido and phenylureido groups), phosphoric acid amido groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly preferably 1 to 12 carbon atoms, such as diethylphosphoric acid amido and phenylphosphoric acid amido groups), a hydroxyl group, a mercapto group, halogen atoms (such as fluorine, chlorine, bromine and iodine atoms), a cyano group, a sulfo group, a carboxyl group, a nitro group, a hydroxamic acid group, an imino group, heterocyclic groups (containing preferably 1 to 30 carbon atoms, more preferably 1 to 12 carbon atoms, and nitrogen, oxygen or/and sulfur atoms as hetero atoms, such as imidazolyl, pyridyl, quinolyl, furyl, thienyl, piperidyl, morpholino, benzoxazolyl, benzimidazolyl and benzothiazolyl groups) and silyl groups (containing preferably 3 to 40 carbon atoms, more preferably 3 to 30 carbon atoms, particularly preferably 3 to 24 carbon atoms, such as trimethylsilyl and triphenylsilyl groups). The substituent groups as recited above may further be substituted.

[0016]

Of the arylene groups as recited above, those preferred as each of Ar^{11} , Ar^{21} and Ar^{31} are phenylene, naphthylene, anthracenylene, phenanthrenylene, pyrenylene, perylenylene and biphenylene groups. Of these groups, phenylene, naphthylene, anthracenylene and pyrenylene groups are much preferred over the others. Further, phenylene and pyrenylene groups, especially pyrenylene group, are advantageous.

[0017]

Ar^{12} , Ar^{22} and Ar^{32} represent a substituent group or a hydrogen atom. Examples of such a substituent group include the groups recited above as substituent groups with which Ar^{11} may be substituted. Preferably, Ar^{12} , Ar^{22} and Ar^{32} are each a hydrogen atom, an aryl group, a heteroaryl group, an alkyl group or an alkenyl group. Of these groups, a hydrogen atom, an aryl group and a heteroaryl group are preferred over the others. Further, it is advantageous for each of Ar^{12} , Ar^{22} and Ar^{32} to be a hydrogen atom or an aryl group, especially a hydrogen atom, an anthracenyl group or a pyrenyl group.

[0018]

At least one of Ar^{11} , Ar^{21} , Ar^{31} , Ar^{12} , Ar^{22} and Ar^{32} has a condensed-ring aryl structure or a condensed-ring heteroaryl structure. It is preferable that at least one of Ar^{11} , Ar^{21} , Ar^{31} , Ar^{12} , Ar^{22} and Ar^{32} has a condensed-ring aryl structure. Suitable examples of such a condensed-ring aryl structure include naphthalene, anthracene, phenanthrene, pyrene and perylene structures. Of these structures, naphthalene, anthracene and pyrene structures are preferred over the others. Further, it is advantageous for the condensed-ring aryl structure to be an anthracene or pyrene structure, especially a pyrene structure.

[0019]

Suitable examples of a condensed-ring heteroaryl

structure include quinoline, quinoxaline, quinazoline, acridine, phenanthridine, phthalazine and phenanthroline structures. Of these structures, quinoline, quinoxaline, quinazoline, phthalazine and phenanthroline structures are preferred over the others.

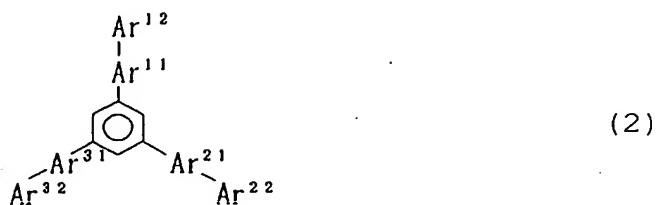
[0020]

Ar represents an arylene group (containing preferably 6 to 30, more preferably 6 to 20, particularly preferably 6 to 16, carbon atoms, such as a phenylene group, a naphthylene group, an anthracenylene group, a phenanthrene group, a pyrenylene group or a triphenylene group), or a heteroarylene group (containing as a hetero atom preferably a nitrogen, sulfur or oxygen atom, more preferably a nitrogen atom, and further containing preferably 2 to 30, more preferably 3 to 20, particularly preferably 3 to 16, carbon atoms, such as a pyridilene group, a pyrazilene group, a thiophenylene group, a quinolilene group or a quinoxalilene group). Each of these groups may have a substituent group. Examples of such a substituent group include the groups recited above as substituent groups Ar¹¹ may have. The group preferred as Ar is a phenylene group, a naphthylene group, an anthracenylene group, a pyrenylene group or a triphenylene group. In particular, and more preferably is a phenylene group, and still more preferably is an unsubstituted phenylene group (Ar¹¹, Ar²¹ and Ar³¹ are substituted).

[0021]

It is preferable for the compound of formula (1) to take a form represented by formula (2). Ar^{11} , Ar^{21} , Ar^{31} , Ar^{12} , Ar^{22} and Ar^{32} in formula (2) have the same meanings as Ar^{11} , Ar^{21} , Ar^{31} , Ar^{12} , Ar^{22} and Ar^{32} in formula (1) respectively.

[0022]



[0023]

The compounds preferred as compounds represented by formula (1) or (2) are compounds in which at least one of Ar^{11} , Ar^{21} , Ar^{31} , Ar^{12} , Ar^{22} and Ar^{32} has a condensed-ring aryl structure in which at least 3 rings are condensed or a condensed-ring heteroaryl structure in which at least 3 rings are condensed, compounds in which Ar^{11} , Ar^{21} and Ar^{31} are condensed-ring arylene groups, or compounds in which Ar^{12} , Ar^{22} and Ar^{32} are a condensed-ring aryl or a condensed-ring heteroaryl group. More preferably, they are compounds in which Ar^{11} , Ar^{21} and Ar^{31} are condensed-ring arylene groups, or compounds in which Ar^{12} , Ar^{22} and Ar^{32} are condensed-ring aryl or heteroaryl groups. Further preferably, they are compounds in which Ar^{11} , Ar^{21} and Ar^{31} are 3 or more rings-condensed arylene groups, or compounds in which Ar^{12} , Ar^{22} and Ar^{32} are 3 or more rings-condensed aryl or heteroaryl

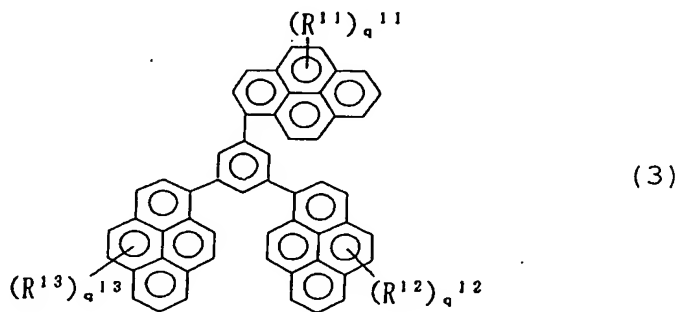
groups. In particular, compounds in which Ar^{11} , Ar^{21} and Ar^{31} are 3 or more rings-condensed arylene groups, or compounds in which Ar^{12} , Ar^{22} and Ar^{32} are 3 or more rings-condensed aryl groups are advantageous over the other compounds.

[0024]

Furthermore, it is advantageous for the compounds represented by formula (1) or (2) to be constituted of carbon and hydrogen atoms alone.

It is preferable for the compound of formula (2) to take a form represented by formula (3).

[0025]



[0026]

In formula (3), R^{11} , R^{12} and R^{13} represent a substituent group. Examples of such a substituent group include the groups recited hereinbefore as substituents which may be present on Ar^{11} . The groups preferred as R^{11} , R^{12} and R^{13} include alkyl, alkenyl, aryl, heteroaryl and alkoxy groups. Of these groups, alkyl and aryl groups, especially aryl groups, are advantageous.

q^{11} , q^{12} and q^{13} are an integer of 0 to 9, preferably 0 to 3, more preferably 0 to 2, particularly preferably 0 or 1.

[0027]

The present compound may be the so-called low molecular compound containing one repeating unit derived from formula (1), or the so-called oligomer or polymer compound containing a plurality of repeating units derived from formula (1) (weight average molecular weight of which is preferably from 1,000 to 5,000,000, more preferably from 2,000 to 1,000,000, particularly preferably from 3,000 to 100,000, on a polystyrene basis). In the case of a polymer compound, the structure represented by formula (1) may be present in the main chain or side chains of the polymer. And the polymer compound may be a homopolymer or copolymer compound. However, the present compound is preferably a low molecular compound.

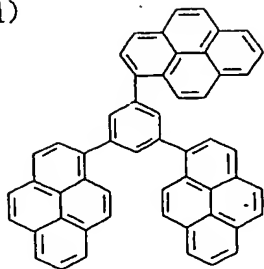
[0028]

Moreover, it is advantageous that the present compound shows the λ_{max} of its fluorescent spectrum (maximum luminescent wavelength) in the region of 400 to 500 nm, preferably 400 to 480 nm, particularly preferably 400 to 460 nm.

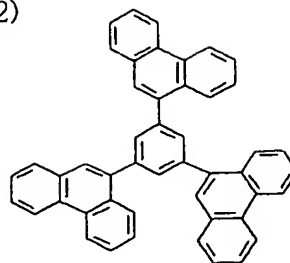
Examples of compounds represented by formulae (1), (2) and (3) are illustrated below, but these examples should not be construed as limiting the scope of the invention in any way.

[0029]

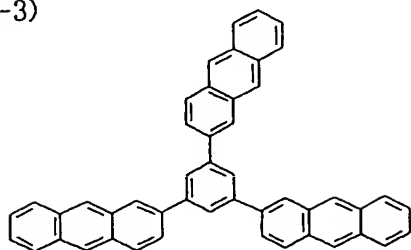
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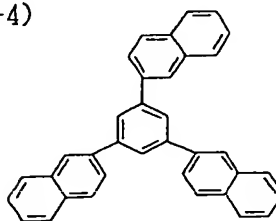
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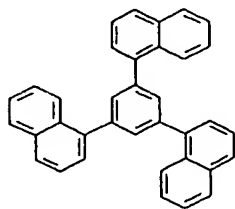
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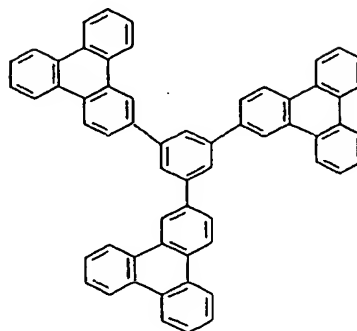
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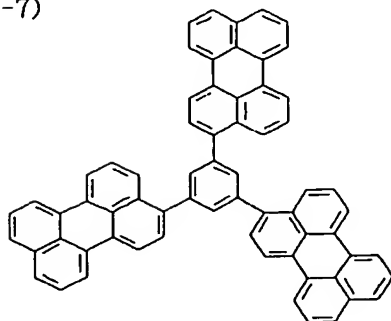
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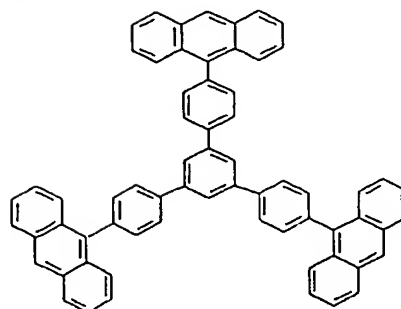
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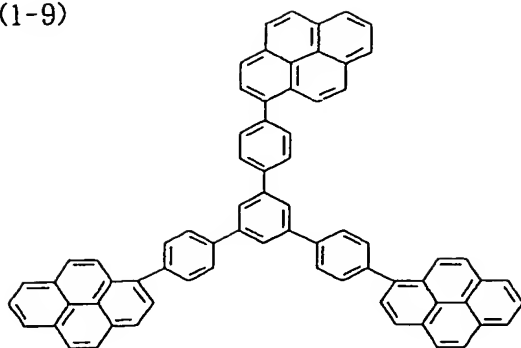


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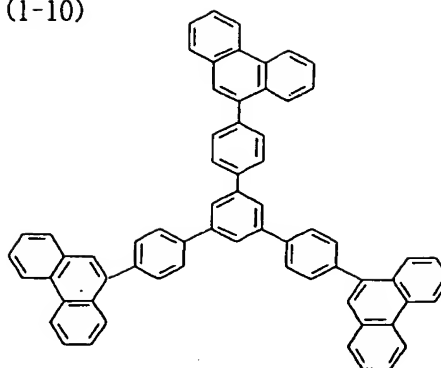


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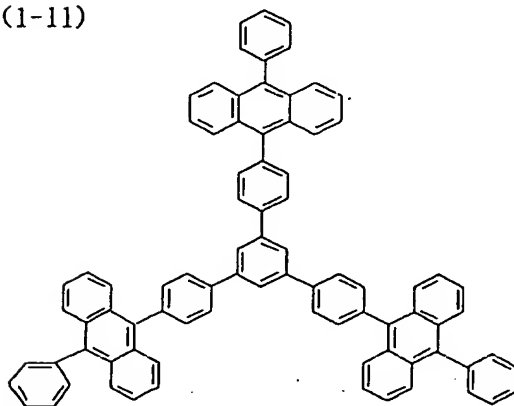
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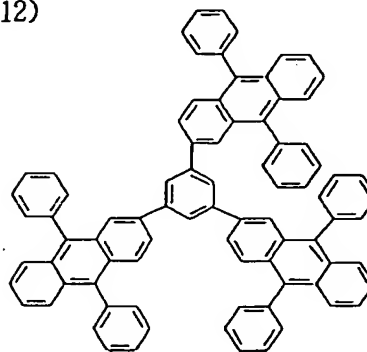
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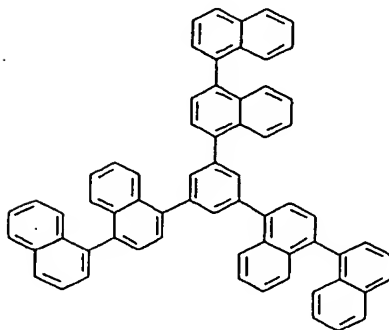
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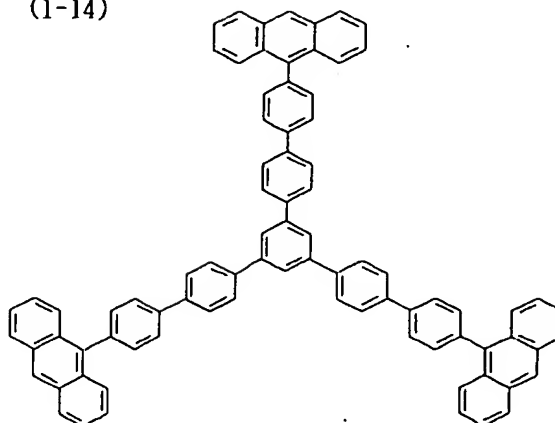
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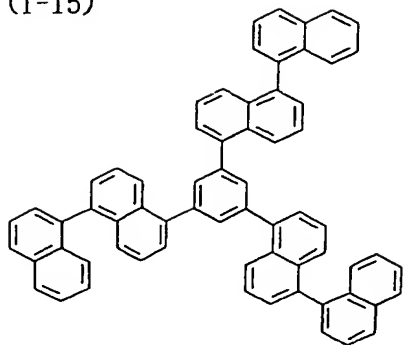


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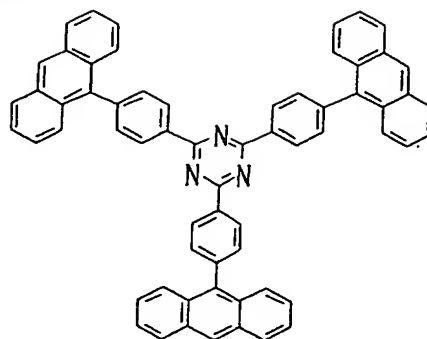


[0031]

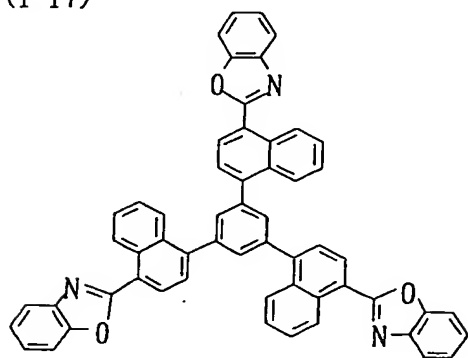
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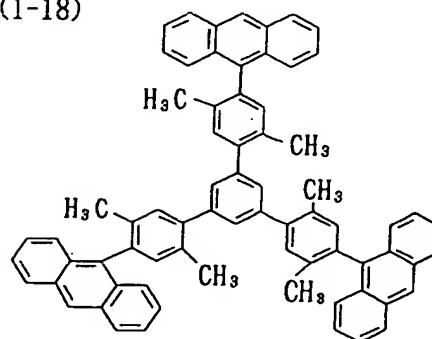
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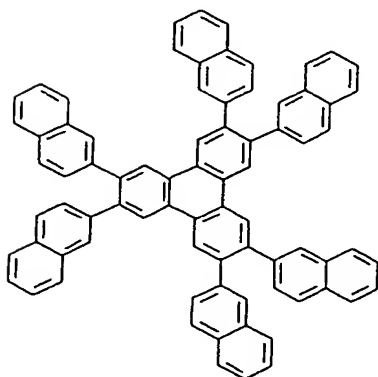
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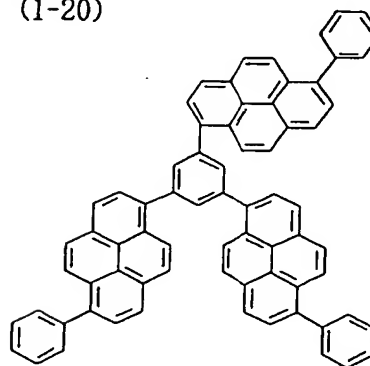
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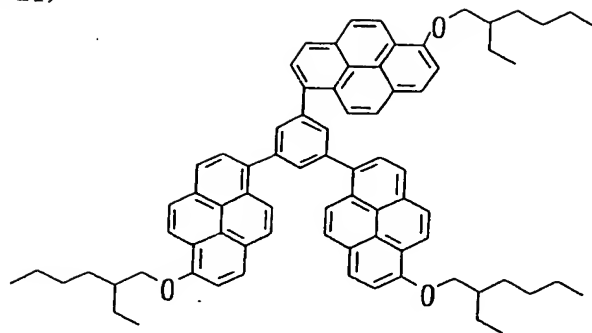


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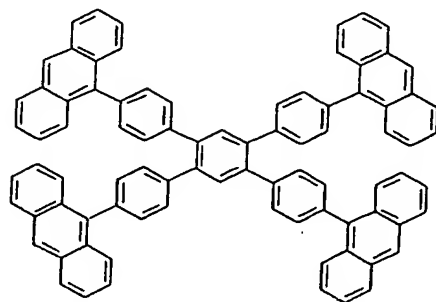


[0032]

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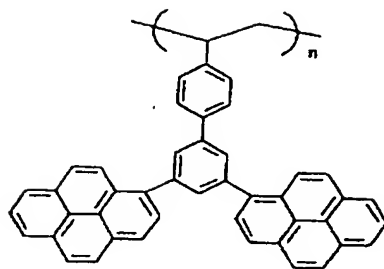


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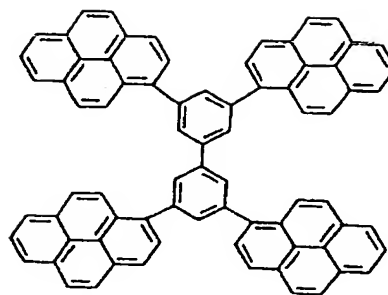
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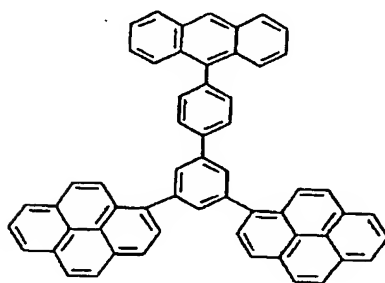


Molecular Weight Mw
(on polystyrene basis): 4,200

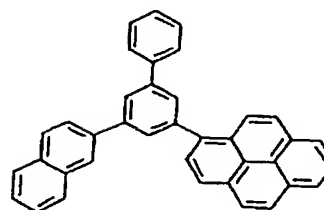
(1-24)



(1-25)



(1-26)



[0034]

In the second place, methods of synthesizing the present compounds are described below.

The present compounds can be synthesized utilizing reactions for forming bonds between aromatic carbons. For the bond formation can be adopted the methods described, e.g., in Organic Synthesis Reaction Guide, pp. 617-643, John Wiley & Sons, Inc., and Comprehensive Organic Transformation, pp. 5-103, VCH Co., Ltd. Of these known methods, the synthesis methods of forming carbon-carbon bonds in the presence of palladium catalysts are preferable. Further, it is advantageous to synthesize the intended compounds by reacting boric acid derivatives with aryl halide derivatives in the presence of palladium catalysts. Examples of such boric acid derivatives include substituted or unsubstituted arylboric acid derivatives (such as benzene-1,4-diboric acid and biphenyl-4,4'-diboric acid) and substituted or unsubstituted heteroarylboric acid derivatives (such as pyridinediboric acid).

[0035]

The halogen atoms of aryl halide derivatives are preferably chlorine, bromine or iodine atoms, particularly preferably bromine atoms.

The foregoing reactions have no particular restrictions as to palladium catalysts. Examples of palladium catalysts

usable therein include palladium tetrakis(triphenylphosphine), palladium carbon, palladium acetate and palladium dichloride (dppf) (dppf: 1,1'-bis(diphenylphosphino)ferrocene). Ligands, such as triphenylphosphine, may be added simultaneously with those palladium catalysts.

In the reactions, it is preferable to use bases. The bases used are not particularly restricted in their kinds, but any of amines, such as sodium carbonate, sodium acetate and triethylamine, can be used. Also, the bases are not particularly restricted in their amounts used. However, the suitable amounts of bases used are from 0.1 to 20 equivalents, preferably from 1 to 10 equivalents, to the boric acid (ester) moiety.

[0036]

In the reactions, it is also preferably to use solvents. Examples of solvents used, though they are not particularly restricted, include ethanol, water, ethylene glycol dimethyl ether, diethylene glycol dimethyl ether, dimethylformamide, toluene, tetrahydrofuran and mixtures of two or more thereof.

Then, light emitting devices comprising the present compounds are illustrated below.

It doesn't matter what system, operation method or utilization mode is adopted by the present light emitting devices so long as the present compounds are used in the devices. However, the devices utilizing luminescence from the present compounds

and the devices utilizing the present compounds as electron transport materials are preferred as the present devices. The representatives of light emitting devices are organic EL (electroluminescence) devices.

The organic layers of the devices containing the present compounds are not particularly restricted in their formation methods, but they can be formed using, e.g., a resistance heating vapor deposition method, an electron beam method, a sputtering method, a molecular lamination method, a coating method or an inkjet method. Of these methods, the resistance heating vapor deposition method and the coating method are preferred in the characteristic and manufacturing aspects.

[0037]

Every light emitting device relating to the invention is a device in which a luminescent layer or at least two thin layers of organic compounds, including a luminescent layer, are formed between a pair of electrodes, namely an anode and a cathode. The thin layers the device may have in addition to the luminescent layer are, e.g., a hole injection layer, a hole transport layer, an electron injection layer, an electron transport layer and a protective layer. Each of these layers may have another function. For forming each layer, various materials can be employed.

The anode supplies holes to a hole injection layer, a hole transport layer and a luminescent layer. As anode

materials, metals, alloys, metal oxides, electrically conductive materials and mixtures thereof, preferably materials having a work function of at least 4 eV, can be used. Examples of such materials include conductive metal oxides, such as tin oxide, zinc oxide, indium oxide and indium tin oxide (ITO), metals such as gold, silver, chromium and nickel, mixtures or laminates of those metals and conductive metal oxides, inorganic conductive materials such as copper iodide and copper sulfide, organic conductive materials such as polyaniline, polythiophene and polypyrrole, and laminates of those materials and ITO. Of the materials recited above, the conductive metal oxides, especially ITO, are advantageous over the others from the viewpoints of productivity, conductivity and transparency. The suitable thickness of the anode, though can be selected depending on the anode material, is generally from 10 nm to 5 μm , preferably 50 nm to 1 μm , particularly preferably 100 nm to 500 nm.

[0038]

In general the anode is used in the state of a layer formed on a soda lime glass, alkali-free glass or transparent resin substrate. In a case of using a glass substrate, alkali-free glass is preferred from the viewpoint of reduction in ions eluted from the glass. When soda glass is used as the substrate, it is desirable that the barrier coat, such as silica, be provided on the glass. The substrate thickness has no particular

limitation so long as the substrate can ensure mechanical strength for the anode. For instance, the suitable thickness of a glass substrate is at least 0.2 mm, preferably at least 0.7 mm.

The methods suitable for making the anode vary with the material used. In the case of ITO, for example, the film formation can be carried out using an electron beam method, a sputtering method, a resistance heating vapor deposition method, a chemical reaction method (e.g., sol-gel method) or the method of coating a dispersion of indium tin oxide.

Washing and other treatments for the anode enable the device to get a reduction in operation potential and improve in light-emitting efficiency. In the case of an anode using ITO, it is effective for the anode to receive UV-ozone treatment or plasma treatment.

[0039]

The cathode supplies electrons to an electron injection layer, an electron transport layer and a luminescent layer. In selecting the cathode, the adhesiveness to the electron injection, electron transport or luminescent layer adjacent to the cathode, the ionization potential and the stability are taken into consideration. As cathode materials, metals, alloys, metal halides, metal oxides, electrically conductive materials and mixtures thereof can be employed. Examples of such materials include alkali metals (e.g., Li, Na, K) and the

fluorides thereof, alkaline earth metals (e.g., Mg, Ca) and the fluorides thereof, gold, silver, lead, aluminum, Na-K alloy or mixtures of two or more of these metals, Li-Al alloy or mixture, Mg-Ag alloy or mixture, and rare earth metals (e.g., In, Yb). Of these materials, the materials having a work function of at most 4 eV are advantageous over the others. In particular, aluminum, Li-Al alloy or mixture and Mg-Ag alloy or mixture are preferably used. The cathode may take a single-layer or made up of the compound or mixture as recited above or a lamination structure comprising the compounds or/and mixtures as recited above. The suitable thickness of the cathode, though can be chosen depending on the cathode material, is generally from 10 nm to 5 μm , preferably 50 nm to 1 μm , particularly preferably 100 nm to 1 μm .

In forming the cathode, various known methods, such as an electron beam method, a sputtering method, a resistance heating vapor deposition method and a coating method, can be adopted. The metals as recited above may be evaporated independently, or two or more thereof may be evaporated simultaneously. Further, it is possible to evaporate a plurality of metals at the same time to form an alloy electrode, or to evaporate the previously prepared alloy.

It is advantageous to the light emitting device that both anode and cathode have low sheet resistance, specifically several hundreds Ω/\square at the highest.

[0040]

Any material can be used as a material for a luminescent layer so far as it can form a layer having a function of receiving both hole injection from the anode, the hole injection layer or the hole transport layer and electron injection from the cathode, the electron injection layer or the electron transport layer when the electric field is applied thereto, a function of permitting the charges injected therein to move and a function of enabling the emission of light by providing a place for recombining the holes and the electrons. Examples of such a material include benzoxazole derivatives, benzimidazole derivatives, benzothiazole derivatives, styrylbenzene derivatives, polyphenyl derivatives, diphenylbutadiene derivatives, tetraphenylbutadiene derivatives, naphthalimide derivatives, coumarin derivatives, perylene derivatives, perinone derivatives, oxadiazole derivatives, aldazine derivatives, pyrazidine derivatives, cyclopentadiene derivatives, bisstyrylanthracene derivatives, quinacridone derivatives, pyrrolopyridine derivatives, thiadiazolo-pyridine derivatives, styrylamine derivatives, aromatic dimethyldyne derivatives, various metal complexes represented by metal or rare earth complexes of 8-quinolinol derivatives, polymeric compounds such as polythiophene, polyphenylene and polyphenylenevinylene, organic silane derivatives, and the present compounds. Although the

luminescent layer has no particular restrictions as to the thickness, the suitable thickness thereof is generally from 1 nm to 5 μ m, preferably 5 nm to 1 μ m, particularly preferably 10 nm to 500 nm.

As to the method of forming the luminescent layer, there is no particular restrictions, but various methods including a resistance heating vapor deposition method, an electron beam method, a sputtering method, a molecular lamination method, a coating method (e.g., a spin coating, cast coating or dip coating method), an inkjet method and an LB method can be adopted. Of these methods, resistance heating vapor deposition and coating methods are favored over the others.

[0041]

The materials for the hole injection layer and the hole transport layer may be any materials so long as they have any one of the functions as an injector of holes from the anode, a transporter of holes and a barrier against electrons injected from the cathode. Examples of materials hitherto known to have one of such functions include carbazole derivatives, triazole derivatives, oxazole derivatives, oxadiazole derivatives, imidazole derivatives, polyarylethane derivatives, pyrazoline derivatives, pyrazolone derivatives, phenylenediamine derivatives, arylamine derivatives, amino-substituted chalcone derivatives, styrylanthracene derivatives, fluorenone derivatives, hydrazone derivatives, stilbene

derivatives, silazane derivatives, aromatic tertiary amine compounds, styrylamine compounds, aromatic dimethyldiyne compounds, porphyrin compounds, polysilane compounds, conductive polymers and oligomers such as poly(N-vinylcarbazole) derivatives, aniline copolymers, thiophene oligomers and polythiophene, organic silane compounds, and the present compounds. The suitable thickness of the hole injection layer and the hole transport layer each, though it has no particular limitation, is generally from 1 nm to 5 μ m, preferably 5 nm to 1 μ m, particularly preferably 10 nm to 500 nm. Each of the hole injection layer and the hole transport layer may have a single-layer structure constituted of one or more of the materials recited above or a multiple-layer structure made up of at least two layers having the same composition or different compositions.

As a method of forming the hole injection or transport layer, a vacuum evaporation method, an LB method, a method of coating a hole-injecting or transporting agent in the form of a solution or dispersion in an appropriate solvent (using, e.g., a spin coating, cast coating or dip coating method) and an ink jet method can be adopted. When the coating method is adopted, the agent to constitute the layer may be dissolved or dispersed in a coating solvent, together with a resinous ingredient. Examples of such a resinous ingredient include polyvinyl chloride, polycarbonate, polystyrene, polymethyl methacrylate,

polybutylmethacrylate, polyester, polysulfone, polyphenylene oxide, polybutadiene, poly(N-vinylcarbazole), hydrocarbon resin, ketone resin, phenoxy resin, polyamide, ethyl cellulose, polyvinyl acetate, ABS resin, polyurethane, melamine resin, unsaturated polyester resin, alkyd resin, epoxy resin and silicone resin.

[0042]

The materials for the electron injection layer and the electron transport layer may be any materials so long as they have any one of the functions as an injector of the electrons from the cathode, a transporter of electrons and a barrier against holes injected from the anode. Examples of compounds known to have such a function include triazole derivatives, oxazole derivatives, oxadiazole derivatives, fluorenone derivatives, anthraquinodimethane derivatives, anthrone derivatives, diphenylquinone derivatives, thiopyran dioxide derivatives, carbodimide derivatives, fluorenylidene methane derivatives, distyrylpyrazine derivatives, tetracarboxylic acid anhydrides of aromatic condensed rings such as naphthalene and perylene, phthalocyanine derivatives, various metal complexes represented by metal complexes of 8-quinolinol derivatives, metallophthalocyanines and metal complexes having benzoxazole or benzothiazole ligands, organic silane derivatives and the present compounds. The suitable thickness of the electron injection layer and the electron transport layer

each, though it has no particular limitation, is generally from 1 nm to 5 μm , preferably 5 nm to 1 μm , particularly preferably 10 nm to 500 nm. Each of the electron injection layer and the electron transport layer may have a single-layer structure constituted of one or more of the materials as recited above, or a multiple-layer structure made up of at least two layers having the same composition or different compositions.

As a method of forming the electron injection layer and the electron transport layer, a vacuum evaporation method, an LB method, a method of coating the electron-injecting or transporting agent as recited above in the form of a solution or dispersion in an appropriate solvent (using, e.g., a spin coating, cast coating or dip coating method) and an ink jet method can be adopted. In the case of adopting a coating method, the electron-injecting or transporting agent can be dissolved or dispersed together with a resinous ingredient. Examples of a resinous ingredient usable therein include the same resins as employed for the hole injection and transport layers.

[0043]

The materials for a protective layer may be any substances so long as they have a function capable of inhibiting the invasion of a device deterioration promoter, such as moisture or oxygen, into the device. Examples of such a substance include metals such as In, Sn, Pb, Au, Cu, Ag, Al, Ti and Ni, metal oxides such as MgO, SiO, SiO₂, Al₂O₃, GeO, NiO, CaO, BaO, Fe₂O₃,

Y₂O₃ and TiO₂, metal fluorides such as MgF₂, LiF, AlF₃ and CaF₂, polyethylene, polypropylene, polymethyl methacrylate, polyimide, polyurea, polytetrafluoroethylene, polychlorotrifluoroethylene, polydichlorodifluoroethylene, copolymer of chlorotrifluoroethylene and dichlorodifluoro-ethylene, copolymers prepared by polymerizing a mixture of tetrafluoroethylene and at least one comonomer, and fluorine-containing copolymers having cyclic structures on the main chain, a water-absorbing substance having a water absorption rate of at least 1 %, and a moistureproof substance having a water absorption rate of at most 0.1 %.

The protective layer also has no particular restriction as to the formation method, but any of a vacuum evaporation method, a sputtering method, a reactive sputtering method, a molecular beam epitaxy (MBE) method, a cluster ion beam method, an ion plating method, a plasma polymerization method (high frequency excitation ion plating method), a plasma chemical vapor deposition (CVD) method, a laser CVD method, a heat CVD method, a gas source CVD method and a coating method can be adopted for the formation thereof.

[0044]

[Examples]

The present invention will now be illustrated in more detail by reference to the following examples. However, the invention should not be construed as being limited to these

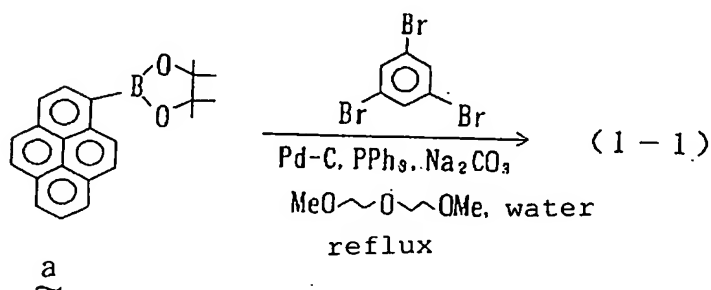
examples.

Synthesis of Compound (1-1)

To a mixture of 1.0 g of pyreneboric acid ester (a), 0.29 g of 1,3,5-tribromobenzene, 0.6 g of sodium carbonate, 0.05 g of triphenylphosphine and 0.05 g of palladium carbon, 20 ml of diethylene glycol dimethyl ether and 20 ml of water were added, followed by stirring under reflux. After 6 hours, the reaction solution was diluted with 200 ml of chloroform and 200 ml of water, and subjected to cerite filtration. The organic layer obtained was washed with 100 ml each of water for two times, dried over sodium sulfate, and then concentrated. The concentrate obtained was purified by column chromatography (chloroform), and further purified by re-crystallization (chloroform/methanol) to yield 0.5 g of Compound (1-1). A vacuum evaporation film of Compound (1-1) was formed, and the fluorescence thereof was measured. The fluorescence maximum wavelength (λ_{max}) of this film was 480 nm.

[0045]

Reaction scheme of Compound (1-1) Synthesis:



[0046]

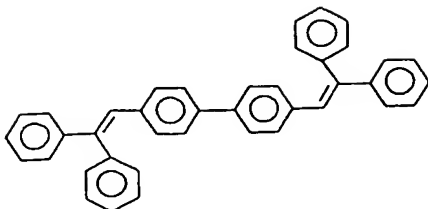
COMPARATIVE EXAMPLE 1

A cleaned ITO substrate was placed in a vacuum evaporation apparatus. Onto this substrate, a 40 nm-thick (N,N-diphenyl-N,N'-di(α -naphthyl)benzidine (α -NPD) film, a 20 nm-thick film of distyryl compound (b) illustrated below and a 40 nm-thick film of azole compound (c) illustrated below were evaporated in order of description. On the thus formed lamination of organic compounds, a patterned mask (for adjusting each emission area to 4 mm \times 5 mm) was set and further, inside the vacuum evaporation apparatus, Mg and Ag were deposited simultaneously in a Mg/Ag ratio of 10/1 to form a metallic film having a thickness of 50 nm, followed by deposition of a 50 nm-thick Ag film.

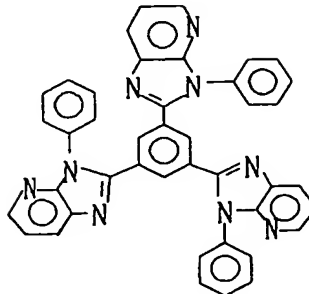
The thus produced EL device was made to luminesce by applying thereto a DC constant voltage by means of a source measure unit, Model 2400, made by Toyo Technica Co., Ltd., and examined for luminance and wavelength of luminescence by using a luminometer BM-8 made by Topcon Co. and a spectrum analyzer PMA-11 made by Hamamatsu Photonics Co. respectively. As a result, it was found that the luminescence was bluish green, the chromaticity value thereof was (0.15, 0.20) and the maximum luminance was 1,130 cd/m². Further, the EL device was allowed to stand for one day in the atmosphere of nitrogen. As a result, white turbidity was observed at the film surface.

[0047]

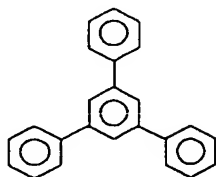
Compound (b)



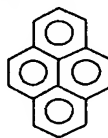
Compound (c)



Compound (d)



Compound (e)



[0048]

COMPARATIVE EXAMPLE 2

An EL device was produced in the same manner as in Comparative Example 1, except that Compound (d) was used in place of Compound (b). There appeared a milky turbidity in the lamination of organic compounds, and so it was impossible to evaluate the device.

COMPARATIVE EXAMPLE 3

An EL device was produced in the same manner as in Comparative Example 1, except that Compound (e) was used in place of Compound (b). There appeared a milky turbidity in the lamination of organic compounds, and so it was impossible

to evaluate the device.

EXAMPLE 1

An EL device was produced in the same manner as in Comparative Example 1, except that the present Compound (1-1) was used in place of Compound (b), and evaluated using the same technique as in Comparative Example 1. As a result, it was found that the luminescence obtained was a bluish green luminescence having a chromaticity value of (0.17, 0.31) and had the maximum luminance of 12,740 cd/m². Even after the device was allowed to stand for one day in the atmosphere of nitrogen, the organic films were transparent.

EXAMPLE 2

An EL device was produced in the same manner as in Comparative Example 1, except that the present Compound (1-8) was used in place of Compound (b), and evaluated using the same technique as in Comparative Example 1. As a result, it was found that the luminescence obtained was a bluish green luminescence having a chromaticity value of (0.16, 0.20) and had the maximum luminance of 6,110 cd/m². Even after the device was allowed to stand for one day in the atmosphere of nitrogen, the organic films were transparent.

EXAMPLE 3

A cleaned ITO substrate was placed in a vacuum evaporation apparatus. Onto this substrate, a 40 nm-thick (N,N-diphenyl-N,N'-di(α -naphthyl)benzidine (α -NPD) film, a 20

nm-thick film of 50:1 mixture of distyryl compound (b) and the present Compound (1-1) and a 40 nm-thick film of azole compound (c) were evaporated in order of description. On the thus formed lamination of organic compounds, the metals were deposited in thin layers in the same manner as in Comparative Example 1. The thus produced EL device was evaporated using the same techniques as in Comparative Example 1. As a result, it was found that the luminescence obtained was a bluish green luminescence having a chromaticity value of (0.16, 0.20) and had the maximum luminance of 11,900 cd/m². Even after the device was allowed to stand for one day in the atmosphere of nitrogen, the organic films were transparent.

EXAMPLE 4

Polyvinylcarbazole in an amount of 40 mg, 12 mg of p-t-butylphenyl-biphenyl-1,2,4-oxadiazole (PBD) and the present Compound (1-21) were dissolved in 3 ml of dichloroethane, and spin-coated on a cleaned substrate (2,000 r.p.m., 5 sec). On this coating, the electrode was deposited in the same manner as Comparative Example 1. The thus produced EL device was evaluated using the same techniques as in Comparative Example 1. As a result, it was found that the luminescence obtained was a bluish green luminescence having a chromaticity value of (0.17, 0.20) and had the maximum luminance of 2,710 cd/m².

EL devices containing other compounds according to the invention were produced and evaluated in the same manner as

described above. Thereby, it was confirmed that the present compounds had high capabilities (luminance, durability and film formability) as an EL device material.

[0049]

[Effect of the Invention]

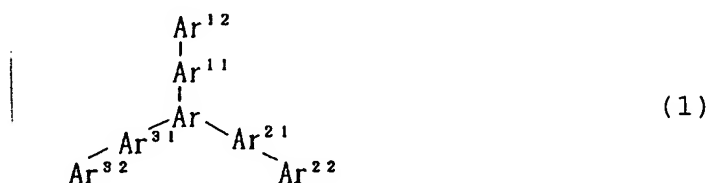
The present compounds are usable as an organic EL material, and besides, they can be used for a medical purpose and also as brightening agents, photographic materials, UV absorbents, laser dyes, color filter dyes and color conversion filters.

[Designation of Document] Abstract

[Abstract]

[Problem] To provide a light emitting device having satisfactory luminescence characteristics and a material enabling a light emitting device to have such characteristics, and a light emitting device using it.

[Means for Resolution] A light emitting device material comprising a compound represented by the following formula (1) is used:



wherein Ar¹¹, Ar²¹ and Ar³¹ represent an arylene group, Ar¹², Ar²² and Ar³² represent a substituent group or a hydrogen atom, provided that at least one of Ar¹¹, Ar²¹, Ar³¹, Ar¹², Ar²² and Ar³² has a condensed-ring aryl structure or a condensed-ring heteroaryl structure, and Ar represents a arylene group or a heteroarylene group.

[Selected Drawing] None